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Lime application lowers the global warming potential of a double rice cropping system

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ABSTRACT

Liming is a common practice to alleviate soil acidification in agricultural systems worldwide. Because liming affects soil microbial activity and soil carbon (C) input rates, it can affect soil greenhouse gas (GHG) emissions as well. However, little is known about the effect of liming on GHG emissions from rice agriculture, one of the main sources of anthropogenic methane (CH_4). Here, we report on the first experiment to measure the effect of liming on GHG emissions from rice paddy fields. We studied a double rice cropping system in an acid paddy for two years and measured the impacts of liming on GHG emissions and rice growth with or without straw incorporation. We found that liming reduced CH_4 emissions in the early rice season, but it did not affect nitrous oxide (N_2O) emissions. Over the two-year study, lime application reduced total CH_4 emissions by 12.5% and 15.4% in plots without and with straw incorporation, respectively. Lime application significantly enhanced rice aboveground biomass, while reducing the area- and yield-scaled global warming potential of CH_4 and N_2O emissions. Lime application stimulated soil respiration during the fallow season and reduced the abundance of methanogens during the early rice growing season. Together, these results suggest that liming reduces CH_4 emissions by promoting the decomposition of organic matter during the fallow season, thereby reducing C availability for methanogens. We conclude that in the short term, liming is an effective practice to reduce greenhouse gas emissions from acidic paddy soils.

1. Introduction

Rice paddies constitute a major source of greenhouse gas (GHG) emissions and account for approximately 20% of global agricultural methane (CH_4) emissions (IPCC, 2013) and 8–11% of China's cropland nitrous oxide (N_2O) emissions (Zou et al., 2009). At the same time, rice is a staple food for about half of the world's population and global demand for rice is expected to increase by 28% in 2050 (Alexandratos and Bruinsma, 2012). Yet, rice yield improvements have recently stagnated in many rice cropping areas (Ray et al., 2012; Grassini et al., 2013). Soil acidity is a main factor limiting rice yield improvement, particularly in subtropical China because of excess N input and the inherent low soil pH (Guo et al., 2010; Miao et al., 2011). Liming is a common management practice for ameliorating soil acidity and increase crop yields, and is commonly applied in rice agriculture (Kirk et al., 2010; Fageria and Nascente, 2014; Holland et al., 2018). However, the effect of lime application on CH_4 and N_2O emissions from rice paddies remains unclear.

Methane is produced by methanogens under anaerobic conditions and is affected by soil pH and C availability (Le Mer and Roger, 2001; Conrad, 2007). Lime application can increase soil pH, which may provide more favorable conditions for CH_4 production (Kunhikrishnan et al., 2016). For instance, Murakami et al. (2005) showed that lime application increased soil pH and then stimulated CH_4 production in peat soils. Liming can also influence soil C availability through improving soil structure, increasing plant productivity, and stimulating soil microbial activity (Paradelo et al., 2015). On the other hand, by increasing soil pH, liming may also stimulate CH_4 oxidation rates (Hilger et al., 2000; Knief et al., 2003). Barton et al. (2014) found that liming increased CH_4 uptake from wheat cropping systems. Thus,

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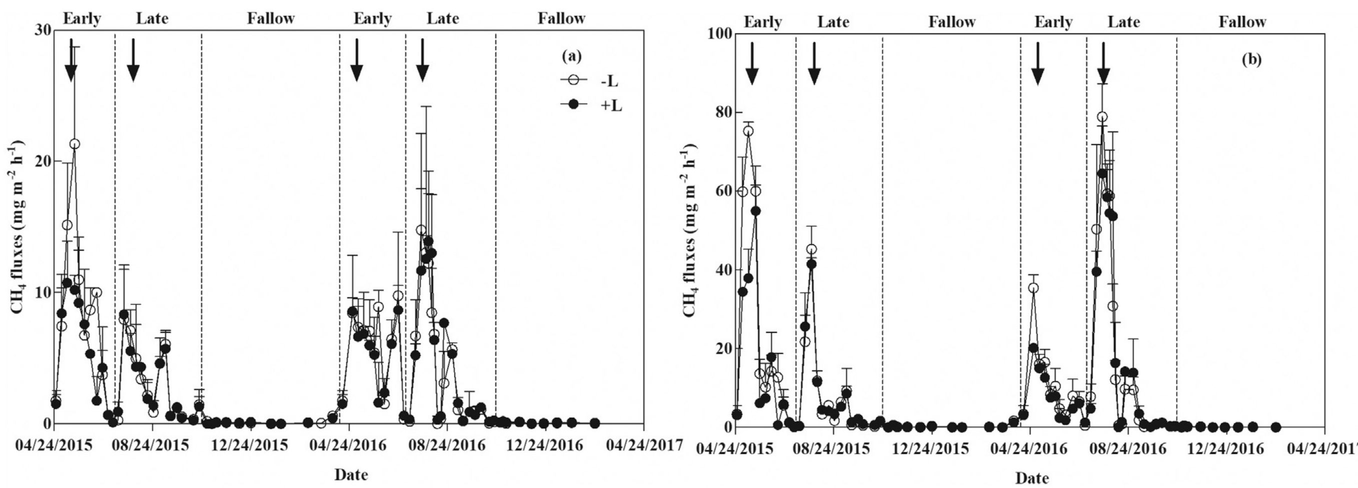

Fig. 1. Effects of liming (−L and +L) on CH_4 fluxes in the plots without (a) and with straw incorporation (b). The arrows indicate the tillering stages. Error bars represent the standard deviation of the mean.

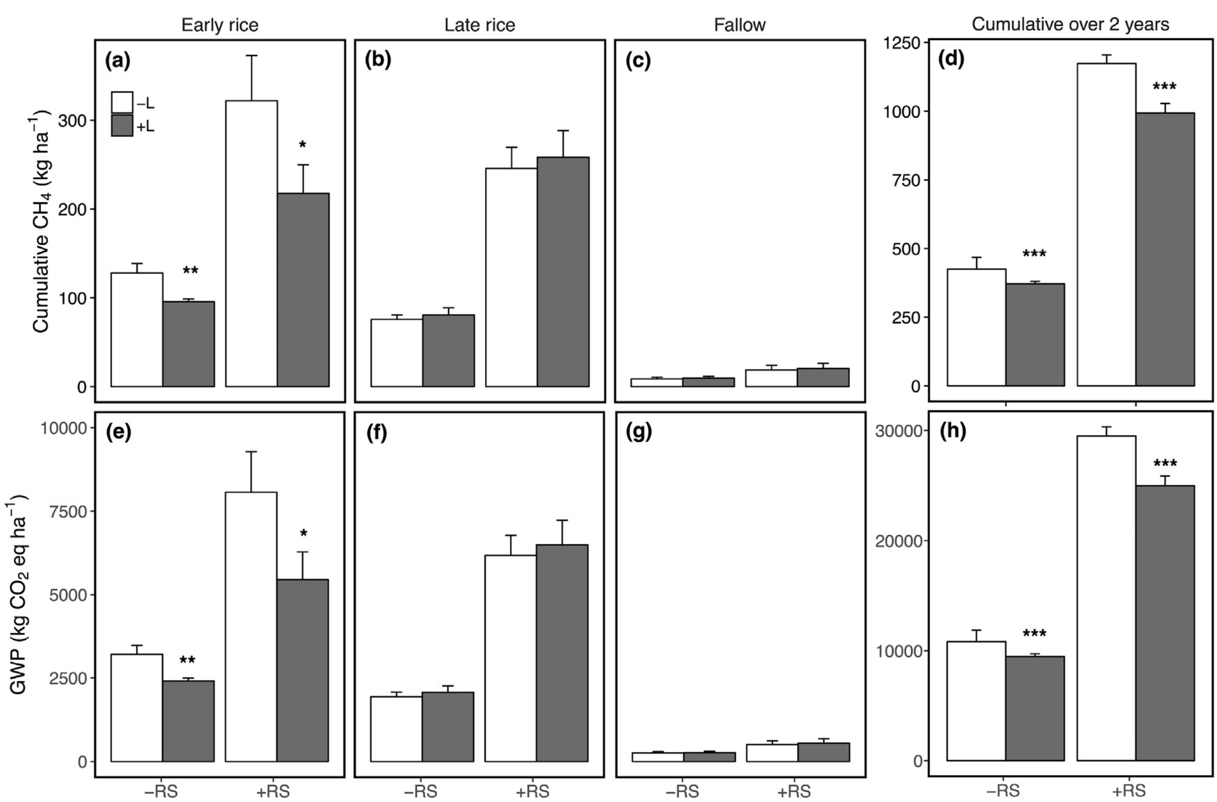

Fig. 2. Seasonal cumulative emissions of CH_4 (a-c) and area-scaled global warming potential (GWP; e–g) during early rice, late rice and fallow seasons in response to liming (−L and +L) and rice straw incorporation (−RS and +RS). Results were averaged across two years because there were no significant 4-way interactions (straw × liming × crop × year). Cumulative CH_4 emissions (d) and GWP (h) for the entire 2-year study period are shown on the right. Error bars represent the standard deviation of the mean. Asterisks (*) indicate significant differences between liming treatments within the straw treatments at $P \leq 0.001$ (***), $0.001 < P \leq 0.01$ (**) or $0.01 < P \leq 0.05$ (*).

evidence suggests that liming can influence both the production, oxidation, and emission of CH_4. However, no experiments have so far assessed the effect of liming on CH_4 emissions from rice paddies.

Soil N_2O is produced mainly through nitrification and denitrification processes (Bouwman, 1998; Zhu et al., 2013). Liming can affect soil nitrification and denitrification rates, thereby influencing N_2O emissions (Kunhikrishnan et al., 2016). For instance, Barton et al. (2013) found that lime application reduced N_2O emission from nitrification. Qu et al. (2014) showed that liming reduced N_2O emissions due to the pH-control of the $N_2O/(N_2O + N_2)$ production ratio. Liming can stimulate plant N uptake (Chang and Sung, 2004), and thereby potentially reduce N availability for denitrification. However, studies measuring the effect of liming on N_2O emissions focused on upland soils; to the best of our knowledge, no study has investigated the impact of liming on N_2O emissions from rice paddy fields.

Besides liming, straw incorporation is another widely applied agricultural practice. Straw incorporation can improve soil fertility, soil organic carbon (SOC) sequestration and rice yield (Huang et al., 2013; Liu et al., 2014). Straw incorporation is applied in approximately 38% of China's rice paddy area, and this percentage is projected to increase

Table 1
Mean cumulative CH_4 and N_2O emissions, area-scaled global warming potential (GWP), yield, biomass, and yield-scaled GWP during the crop growing season as affected by liming (−L and +L), straw retention (−RS and +RS), crop (early and late rice), and study year. *F*-values after model simplification are provided for interactions.

	CH_4 (kg ha^{-1})	N_2O (g ha^{-1})	GWP (kg CO_2 eq ha^{-1})	Biomass (kg ha^{-1})	Yield-scaled GWP (kg CO_2 eq kg^{-1})
Liming (L)[a]					
−L	193	89	4848	12,955	0.73
+L	163***	91	4105***	14,419***	0.51***
Straw (S)[b]					
−RS	95	108	2407	12,906	0.36
+RS	261***	72	6546***	14,467***	0.88***
Crop (C)[c]					
Early	191	49	4785	12,731	0.74
Late	165***	131***	4168***	14,642***	0.51***
Year (Y)[d]					
2015	186	101	4689	14,614	0.66
2016	170**	78	4264***	12,759***	0.58***
F-values					
S × L	7.85*	NS	8.12*	9.76*	27.4***
S × C	2.25	NS	2.05	0.002	0.43
L × C	53.2***	NS	52.0***	14.7***	30.4***
S × Y	6.29*	5.32*	6.19*	10.3**	15.8***
L × Y	13.6**	NS	12.7**	7.16*	5.00*
C × Y	328***	NS	321***	22.5***	201***
S × L × C	14.1***	NS	13.7***	14.1***	5.67*
S × C × Y	196***	NS	190***	6.98*	146***
L × C × Y	5.09*	NS	6.10*	NS	6.21*

There were no significant four-way interactions and one non-significant three-way interaction (S × L × Y) for the six variables. Variables for which the final model no longer contained the non-significant (NS) three-way interactions show NS. Significant treatment effects within a main category are indicated by * ($0.01 < P \leq 0.05$), ** ($0.001 < P \leq 0.01$), or *** ($P \leq 0.001$).
Note that N_2O emissions are in different units than CH_4 emissions.
[a] Values were averaged across straw treatments, crop, and years.
[b] Values were averaged across liming treatments, crop, and years.
[c] Values were averaged across years, straw, and liming treatments.
[d] Values were averaged across crop, straw, and liming treatments.

further (G. Zhang et al., 2017). Because straw incorporation increases substrate availability to methanogens, it typically increases CH_4 emissions from rice paddies (Liu et al., 2014). Liming and straw incorporation affect soil microbes and plant growth through different mechanisms. Thus, these two management practices may interact in their effects on GHG emissions. However, the possible interactions between liming and straw incorporation on the response of GHG emissions have not yet been studied.

The double rice cropping system is one of the most important cropping systems in the world, accounting for approximately 40% of the total rice planting area in China (MOA, 2017). Double rice cropping systems also have the highest GHG emissions among all cereal cropping systems (Linquist et al., 2012; Feng et al., 2013). The double rice cropping system typically consists of early rice (grown from April to July) and late rice (grown from July to November), followed by a 5-month long fallow season during which soils are not flooded. Increasing evidence shows that liming can promote the decomposition of organic matter in unflooded soils (Paradelo et al., 2015; Kunhikrishnan et al., 2016), potentially reducing the amount of substrate available to methanogens following the rice growth season. Thus, we hypothesized that 1) liming mitigates CH_4 emissions, especially in early season, through accelerating the decomposition of crop residues during the fallow season; 2) based on published results from experiments in upland ecosystems, liming decreases N_2O emissions; and 3) liming increases rice yield while mitigating the global warming potential (GWP) of CH_4 and N_2O emissions. To test these hypotheses, we conducted a two-year field experiment to investigate the effects of liming on CH_4 and N_2O emissions and rice yield in a double rice cropping system. To the best of our knowledge, this is the first study to report on the effects of liming on GHG emissions from rice paddies.

2. Materials and methods

2.1. Experimental site

We conducted a field experiment at Zengjia village (115°09′ E, 28°31′ N), Shanggao County, Jiangxi Province, China from 2015 to 2017. This site has a sub-tropical monsoon climate with a mean annual precipitation of 1650 mm and mean annual temperature of 17.5 °C. The soil at our site is a Typic Stagnic Anthrosol. The main soil properties (0–15 cm) are as follows: pH (1:2.5, soil:H_2O) 5.2, organic matter 18.1 g kg^{-1}, total N 1.1 g kg^{-1}, total P 0.37 g kg^{-1}, total K 3.88 g kg^{-1}, alkaline hydrolyzable-N 115.0 mg kg^{-1}, available P 15.9 mg kg^{-1}, and available K 64.0 mg kg^{-1}.

2.2. Experimental design

Our field experiment consisted of twelve plots (5 × 5 m) and had a completely randomized block design, with two levels of liming, two levels of straw management, and three replicates for each treatment combination. Lime application treatments consisted of liming (+L) and no liming (−L), and straw management treatments consisted of rice straw incorporation (+RS) and no straw incorporation (−RS). In the +L plots, $Ca(OH)_2$ was broadcast once in the 2014–2015 fallow season at a rate of 2.1 t ha^{-1}. In the +RS treatments, aboveground rice straw was chopped into 10-cm long pieces. After early rice harvest, rice straw was incorporated into the soil by plowing. After the late rice harvest, the straw was left on the soil surface until the following year, when it was incorporated into the soil by plowing prior to transplanting the early rice seedlings. The 10-cm residual stubbles were left in all plots.

The rice cultivars ‘Zhongjiazao 17’ and ‘Wuyou 308’ were grown as the early and late rice, respectively. The early rice seedling was transplanted at a hill spacing of 13.2 cm × 23.1 cm with four seedlings per

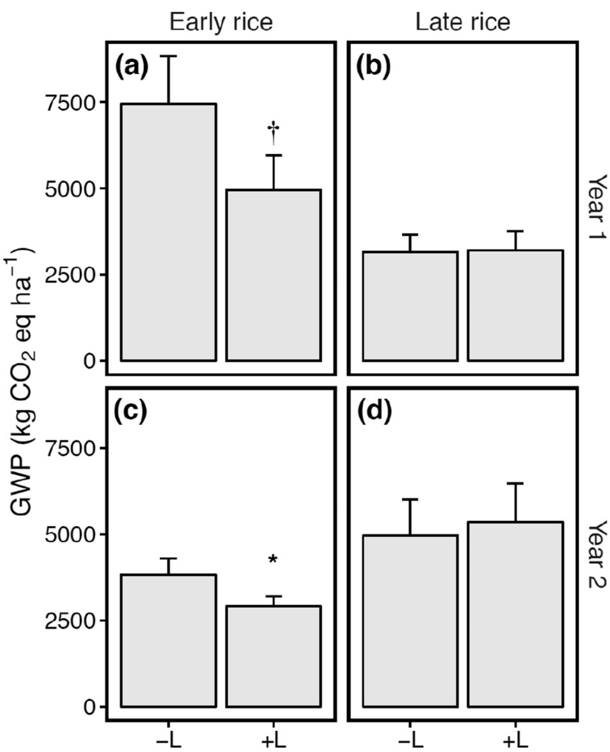

Fig. 3. Effects of liming (−L and +L) on area-scaled global warming potential (GWP) on early (a, c) and late (b, d) seasons in 1st (a, b) and 2nd (c, d) years. GWP for each liming treatment was calculated by pooling across straw treatments. Error bars represent the standard deviation of the mean. ⁺ indicates marginally significant differences between liming treatments at $0.05 < P < 0.1$. * indicate significant differences between liming treatments at $0.01 < P \leq 0.05$.

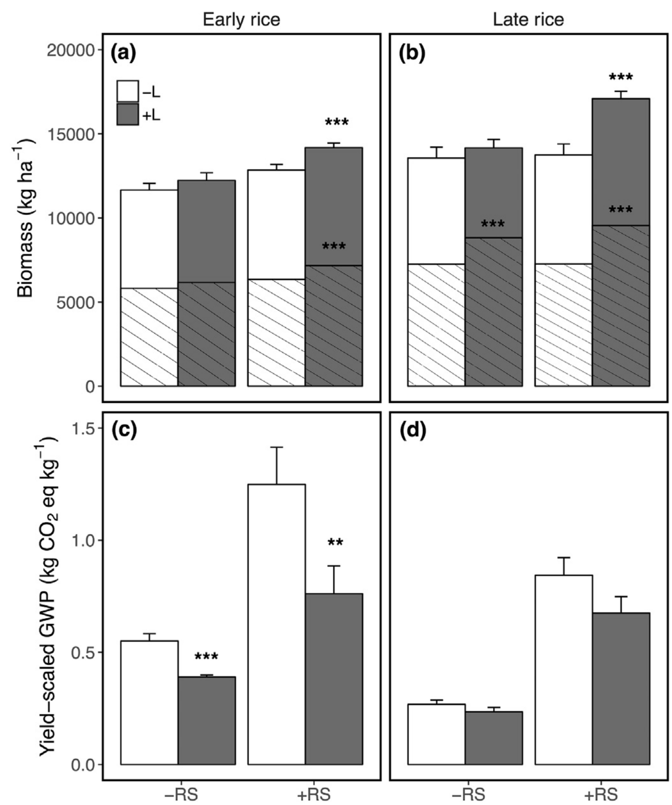

Fig. 4. Aboveground biomass (a, b) and yield-scaled global warming potential (b, c) as affected by liming (−L and +L), rice straw incorporation (−RS and +RS) and their interaction. Error bars represent the standard deviation of the mean. The hatched bars denote rice yields. Asterisks (*) indicate significant differences between liming treatments within the straw treatments at $P \leq 0.001$ (***), $0.001 < P \leq 0.01$ (**) or $0.01 < P \leq 0.05$ (*).

hill on 24 April 2015 and on 21 April 2016. We transplanted the late rice seedling at a hill spacing of 13.2 cm × 26.4 cm with two seedlings per hill on 22 July 2015 and 20 July 2016. The 5-month fallow season started following the harvest of late rice on October 28, 2015 and on October 20, 2016. Nitrogen fertilizer was applied as urea, at 120 kg N ha^{-1} and 150 kg N ha^{-1} in early and late rice seasons, respectively, 50% of which was applied as a basal fertilizer before planting, another 20% at the tillering stage and the remaining 30% at the jointing stage. Phosphorus fertilizer as calcium magnesium phosphate was applied before transplanting of rice seedlings at the rate of 33 kg P ha^{-1} in each season. Potassium fertilizer as potassium chloride was applied at the rate of 31 kg K ha^{-1} before transplanting and at the jointing stage. Water regimes in all plots consisted of early flooding-mid season drainage-intermittent irrigation. During the fallow season, the paddy was drained. Averaged across the rice growing season, the depth of the water layer was approximately 4 cm in all treatments (Fig. S1). All other management practices followed local recommendations.

2.3. Sampling and measurement

We measured CH_4 and N_2O emissions using the static closed chamber technique at one-week intervals during the rice growth seasons, and at two-week intervals during fallow seasons (Jiang et al., 2016). In each plot, we inserted a PVC frame (inner length 50 cm, width 50 cm, and height 15 cm) into the soil at about 3 days after seedling transplanting in rice growth seasons. Each PVC frame included six rice plants. At the same time, we set up the wooden boardwalks in each plot to prevent soil disturbance during sampling. The chambers were temporarily installed on the frame while sampling. The dimensions of the chamber were 50 × 50 × 50/100 cm (L × W × H); chamber height was increased during the growing season to accommodate increased plant height. On each sampling day between 09:00 to 11:00 a.m., we collected gas samples at 5 minute intervals for a total period of 20 min. We used a gas chromatograph (Agilent 7890A, USA) to measure CH_4 and N_2O concentrations. The CH_4 or N_2O fluxes (F) were calculated as follows:

$$F = \Delta C/\Delta T \times V/A,$$

where $\Delta C/\Delta T$ is the change in gas concentration (mg L^{-1} h^{-1}) in the chamber (calculated using linear regression), V is the volume of the chamber (L) and A is enclosed surface area (m^2). Cumulative CH_4 or N_2O emissions were calculated for each season and for the entire study period using linear interpolation using the 'zoo' package in R (v3.3.2; Zeileis and Grothendieck, 2005). The combined 100-year GWP of the CH_4 and N_2O emissions was calculated as follows:

$$\mathrm{GWP}\ (\mathrm{kg\ CO_2\ eq\ ha^{-1}}) = 298 \times \mathrm{N_2O}\ (\mathrm{kg\ N_2O\ ha^{-1}}) + 25 \times \mathrm{CH_4}\ (\mathrm{kg\ CH_4\ ha^{-1}}).$$

Soil respiration during the fallow season was also measured by the static closed chamber technique at two-week intervals between 09:00 to 11:00 a.m.

We collected soil samples (0–15 cm) to measure the methanogenic abundance at the tillering stage of early rice season in 2016, when CH_4 emissions were relatively high and significantly different between treatments. Soil DNA was extracted from 0.25 g soil using a Power Soil DNA Isolation Kit (MoBio, USA). The copy numbers of *mcr*A genes and *pmo*A genes, which represent the abundances of methanogenic archaea and methanotrophic bacteria in soil, were quantified using the primer pair mcrAf/mcrAr and A189f/A682r, respectively (Holmes et al., 1995; Luton et al., 2002). Quantitative real-time PCR was performed using

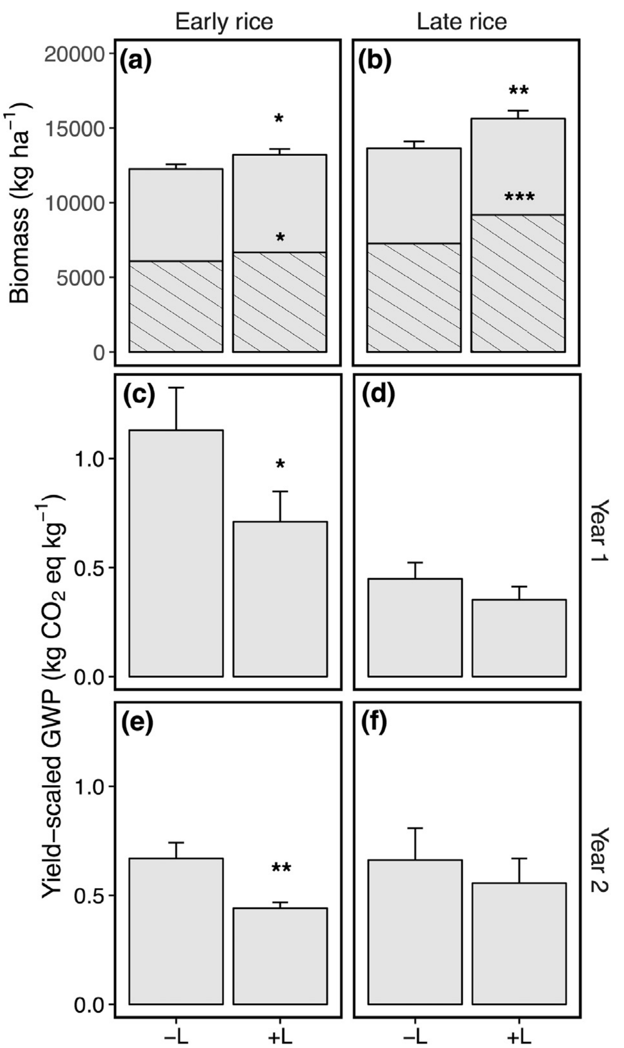

Fig. 5. Effects of liming (−L and +L) on aboveground biomass (a, b) and yield-scaled global warming potential (c–f) on early (a, c, and e) and late (b, d, and f) seasons in 1st (c, d) and 2nd (e, f) years. Yield data are averaged across years, as there was no significant liming × crop × year interaction. Means for each liming treatment were calculated by pooling across straw treatments. Error bars represent the standard deviation of the mean. The hatched bars denote rice yields. Asterisks (*) indicate significant differences between liming treatments at $P \leq 0.001$ (***), $0.001 < P \leq 0.01$ (**) or $0.01 < P \leq 0.05$ (*).

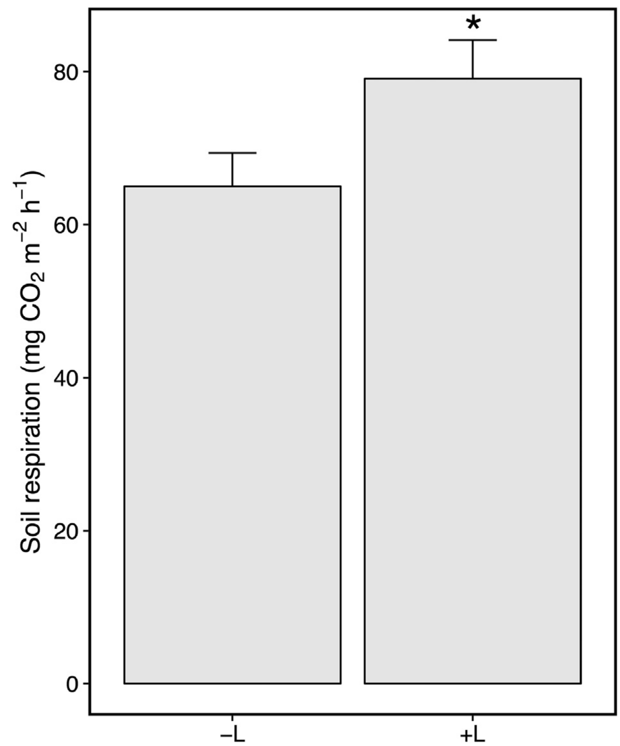

Fig. 6. Mean soil respiration during the fallow seasons of 2015–2016 and 2016–2017 with no liming (−L) and liming (+L). Values were averaged across years and straw treatments. Error bars represent the standard deviation of the mean. The asterisk (*) indicates significant differences between liming treatments at $0.01 < P \leq 0.05$.

Table 2
Mean soil properties and mixed-model *F*-values during the fallow period in 2017 as affected by liming (−L and +L) and straw retention (−RS and +RS). Standard deviations of the mean are shown in parentheses.

Straw	Liming	pH	SOC (g kg^{-1})	Al^{3+} (cmol kg^{-1})
−R	−L	5.27 (0.09)	10.69 (0.21)	1.39 (0.21)
	+L	6.09 (0.03)	11.40 (0.55)	0.75 (0.05)
+R	−L	5.48 (0.21)	12.63 (0.40)	1.20 (0.05)
	+L	6.17 (0.17)	13.73 (0.95)	0.63 (0.03)
F-values				
L		80.1***	102***	7.41*
S		3.01	6.33*	41.5***

There were no significant pairwise interactions between straw and liming (S × L) for soil properties. Significant treatment effects within a treatment are indicated by * ($0.01 < P \leq 0.05$), ** ($0.001 < P \leq 0.01$), or *** ($P \leq 0.001$).

CFX Connect (Bio-Rad, America) and the data were analyzed using CFX Manager System software (Bio-Rad).

We also collected soil samples (0–15 cm) at the 2016 late rice harvest to measure soil pH, SOC, and soil exchangeable Al^{3+}concentrations. Soil pH (1:2.5 H_2O) was measured by a pH meter and SOC was measured by the potassium dichromate oxidation method (Lu, 2000). Soil exchangeable Al^{3+} concentrations were measured by the Sokołow method (Lu, 2000).

For the aboveground biomass measurement, 12 hills were sampled at maturity and dried at 70 °C to a constant weight. Grain yield was determined from a 5 m^2 sampling area in each plot and then was adjusted to a moisture content of 0.14 g H_2O g^{-1} fresh weight. Yield data for our experiment are reported in detail by Liao et al. (2018).

2.4. Statistical analysis

We fitted linear mixed-effects models from the 'nlme' package (Pinheiro et al., 2016) to determine the effects of liming, straw incorporation, year, and their interactions on rice yield, biomass, cumulative CH_4 and N_2O emissions, GWP and soil respiration rates. We used liming, straw, rice crop and year, and all interactions as main effects in the model and because of repeated sampling, we used plot as a random effect. We then used a model simplification procedure by iteratively removing higher order interactions and assessing model performance after each removal (Crawley, 2013). Our mixed-model *F*-values for the response variables are based on the resulting parsimonious models. We used bootstrapping techniques to generate standard deviations of the means (n = 5000 iterations). All analyses were conducted in R (v3.3.2; R Core Team, 2016).

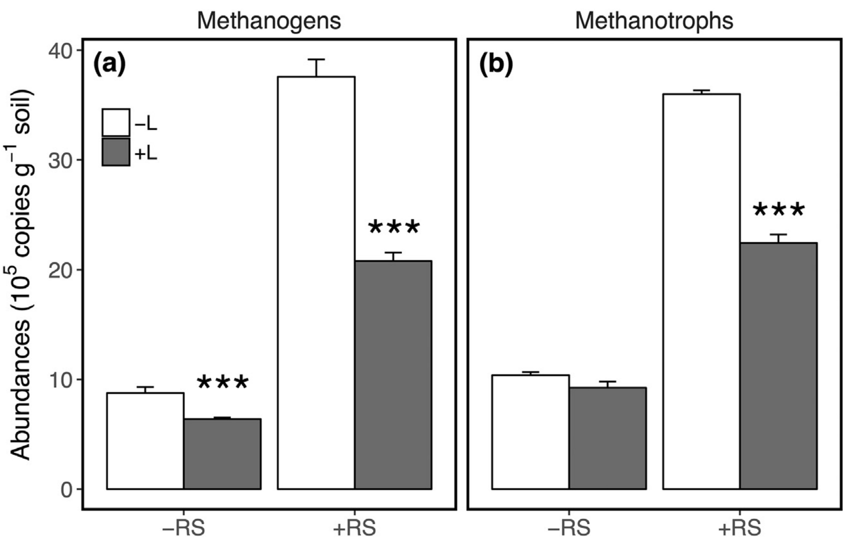

Fig. 7. Abundances of methanogens (a) and methanotrophs (b) during the 2016 early rice season in response to liming (−L and +L), straw treatments (−RS and +RS) and their interaction. Error bars represent the standard deviation of the mean. Asterisks (*) indicate significant differences between liming treatments within the straw treatments at $P \leq 0.001$ (***), $0.001 < P \leq 0.01$ (**) or $0.01 < P \leq 0.05$ (*).

3. Results

3.1. CH_4 emissions

The dynamic of CH_4 fluxes were similar among rice seasons and treatments in that the peak emissions of CH_4 fluxes occurred at the tillering stage in both rice seasons (Fig. 1). Averaged across years and straw treatments, liming reduced cumulative seasonal CH_4 emissions by 30.4% during the early rice seasons (Fig. 2a), but had no effect on CH_4 emissions from the late rice seasons and fallow seasons (Fig. 2b,c). The significant reductions in CH_4 emissions in response to liming during the early rice seasons were substantial enough to reduce cumulative emissions for the entire study period by 12.5% and 15.4% in plots without and with straw incorporation, respectively (Fig. 2d). In both lime management practices, straw incorporation significantly increased CH_4 emissions in all rice seasons and fallow seasons. Pooled across liming treatments, straw incorporation nearly tripled (2.7-fold) cumulative CH_4 emissions over the two-year study. Liming and straw incorporation had an interactive effect (Table 1) on cumulative CH_4 emissions in the early rice season, with liming decreasing emissions more strongly when straw was incorporated (32.4% emission reduction) compared to when straw was absent (−25.3%; Fig. 2a). Liming did not alter how straw affected CH_4 emissions during the late rice or fallow seasons. Among the four treatments, the +RS − L treatment resulted in the highest CH_4 emissions. The effect of liming on CH_4 emission during the early rice growing season was stronger in the first year, with CH_4 emissions reduced by 33.7% in response to liming in the first year and 24.1% in the second year (Table S1).

3.2. N_2O emissions

During the two years, N_2O emissions were highly variable and very low ($< 1\ kg\ ha^{-1}$), and hence, no clear trend of N_2O fluxes was observed in response to liming, straw incorporation or year (Fig. S2; Tables 1 and S1). However, seasonal cumulative N_2O emissions were larger in the late rice season than in the early rice season (Tables 1 and S1).

3.3. Area-scaled GWP

Because CH_4 emissions contributed at least 96% to area-scaled GWP values, N_2O emissions had minimal effects on GWP patterns (Fig. 2, Tables 1 and S1). Liming reduced area-scaled GWP with or without straw incorporation in the early rice seasons by 30.3% on average for both years (Figs. 2e and 3a,c), but did not affect area-scaled GWP in the late rice seasons and fallow seasons (Figs. 2f,g and 3b,d). Nevertheless, as was the case with the CH_4 emissions, the substantial reductions in the early rice seasons were sufficient to reduce cumulative GWP for the entire 2-year study period by 14.6% (Fig. 2h). Nearly identical to CH_4 emissions, the effect of liming on GWP was stronger in the first early rice season, with GWP reduced by 33.5% in response to liming in the first early rice season and 24.2% in the second early rice season (Fig. 3). Straw incorporation increased area-scaled GWP during all seasons (Table 1). The +RS − L treatment had the highest area-scaled GWP (Fig. 2h).

3.4. Biomass, yield and yield-scaled GWP

Straw incorporation and lime application both increased aboveground biomass and rice yield (Figs. 4a,b and 5a,b; Tables 1 and S1), but the treatment effects differed between early rice and late rice; liming increased early-rice biomass to a smaller degree (+9.6%) than late-rice biomass (+26.5%), whereas straw incorporation increased the biomass of early rice by 12.8% but not late rice. Similar interactions were found for rice yield (Table S1). Liming and straw incorporation had a positive interactive effect on aboveground biomass, whereby liming increased biomass more strongly in plots with straw incorporation (+22.8%) compared to plots with no straw (+14.6%). Similar interactions were found for treatment effects on rice yield (Table S1). Among the four treatments, the +RS + L treatment had highest aboveground biomass and rice yield (Fig. 3a and Table S1).

Liming reduced yield-scaled GWP in all rice seasons (Figs. 4cd and 5c–f). Averaged across both years, liming reduced GWP by 36.0% during the early rice season. Liming caused significantly smaller reductions in GWP during the late rice seasons (−18.1% on average). Liming had a significant interaction with straw incorporation that differed between the two rice crops (Table 1). Liming without straw incorporation resulted in a 29.1% decrease in yield-scaled GWP in early rice, but with straw incorporation the reduction was stronger, 39.0% (Fig. 4c, Table S1). For late rice, liming reduced yield-scaled GWP only when straw was incorporated (−19.9%; Fig. 4d). The effect of liming on yield-scaled GWP was stronger in the first early rice season than the second early rice season (Fig. 5c,e). Straw incorporation increased yield-scaled GWP during all seasons (Fig. 4c,d; Tables 1 and S1). The +RS − L treatment had the highest yield-scaled GWP (Fig. 4c,d).

3.5. Soil properties, soil respiration, and abundances of methanogens and methanotrophs

Liming significantly increased soil pH and SOC contents (Table 2).

In contrast, liming significantly reduced soil exchangeable Al^{3+} concentrations. Straw incorporation increased SOC contents and decreased soil exchangeable Al^{3+} concentrations. Straw incorporation did not affect soil pH. There were no significant liming × straw interactions for any of these variables.

Lime application and straw incorporation separately affected soil respiration during both fallow seasons (Figs. 6 and S3, $P < 0.001$), with mean soil respiration rates increasing by 21.7% in response to liming and by 43.9% in response to straw incorporation. There were no interactive effects between liming and straw additions for either fallow season.

Liming, straw incorporation and their interaction had significant effects on abundances of methanogens and methanotrophs (all $P < 0.001$), which showed very similar responses (Fig. 7). Averaged across treatments, liming reduced methanogenic abundances by 41.3% and methanotrophic abundances by 31.7%. In contrast, straw incorporation significantly increased methanogenic and methanotrophic abundances. Liming reduced methanogen abundances without straw incorporation (−26.9%); with straw incorporation, liming reduced methanogen abundances to a larger degree, by 44.7%. Methanotrophs did not respond to liming in the absence of straw, but decreased abundances by 37.6% when liming was combined with straw incorporation.

4. Discussion

Liming reduced CH_4 emissions during the early rice season and this reduction coincided with lower methanogenic abundances. Because methanogenic growth is limited by C availability (Conrad, 2007), these results suggest that the effect of liming may have been modulated through its effect on soil C availability. We propose that liming stimulated microbial activity and organic matter decomposition during the fallow season, resulting in reduced C availability for methanogens during the following rice growth season. This explanation is consistent with a recent study conducted at the same site (Liao et al., 2018), showing that liming stimulated the activity of soil enzymes (i.e., invertase and cellulose) involved in soil C cycling. It is also consistent with our finding that liming increased soil respiration during the fallow season, and with numerous studies showing that liming can stimulate soil carbon mineralization in non-flooded soils (e.g. Paradelo et al., 2015). Moreover, liming increased rice plant growth, which may stimulate oxygen transport into rhizosphere during the growing season when soils are flooded (Jiang et al., 2017). Higher oxygen concentrations can inhibit methanogenic growth and methane production from flooded soils (Schrope et al., 1999; Conrad, 2007).

The decrease in methanotrophic abundance with liming could be related to substrate dependency (e.g. Cai et al., 2016); reduced methanogenic activity in limed plots likely lowered CH_4 availability to methanotrophs. However, liming decreased soil methanotrophic abundances to a smaller degree than methanogenic abundances, suggesting that other mechanisms modulated the methanotrophic response to liming. For instance, increased oxygen transport into the rhizosphere with liming (see above) may stimulate methanotrophic growth and CH_4 oxidation (Yuan et al., 2009; Ma and Lu, 2010; Jiang et al., 2017). Moreover, the increase in soil pH with liming reduced the availability of aluminum in our experiment, a response which may stimulate methanotrophic activity as well (Hilger et al., 2000; Knief et al., 2003; Kunhikrishnan et al., 2016).

Why does liming only reduce CH_4 emissions during the early rice growing season? After the early rice harvest, plant residues were immediately incorporated into the soil, which was subsequently flooded prior to late rice planting. The flooded conditions and short time period between the early and late growing seasons likely limited the negative effect of liming on methanogen substrate availability through its positive effect on decomposition rates (Liao et al., 2018). Furthermore, by stimulating rice biomass, liming likely increased soil C input rates and substrate availability for methanogens during the late rice season, negating any negative effects related to increased decomposition rates.

Contrary to our second hypothesis, liming did not affect N_2O emissions. The lack of treatment effects on N_2O emissions is probably related to low O_2 concentrations in rice paddies limiting nitrification rates (Cai et al., 1997). Soil N_2O is produced mainly through the microbial processes of nitrification and denitrification; nitrification dominates soil N_2O production at soil aeration in the range of 30–60% water-filled pore space (WFPS), whereas denitrification is a major process at 50–90% WFPS (Bouwman, 1998). We applied the N as urea (ammonium fertilizer) after flooding, when WFPS is high and O_2 availability is low. Thus, nitrification during the growing season was likely limited, resulting in low production rates of NO_3^-, which in turn inhibited denitrification. Similarly, our finding that straw incorporation did not affect soil N_2O emissions either is probably related to low O_2 concentrations. These results corroborated several other studies (Hang et al., 2014; J. Zhang et al., 2017). A global meta-analysis also showed that crop residue addition did not affect N_2O emissions when WFPS is > 90% (Chen et al., 2013), as is the case in flooded soils.

In agreement with previous studies, liming significantly increased rice biomass (e.g. Chang and Sung, 2004; Crusciol et al., 2016). Furthermore, we found that lime addition increased rice biomass more strongly in treatments with straw addition. This may be because liming can neutralize organic acids released by the anaerobic decomposition of straw, which can offset the negative effects of organic acid accumulation on rice plant growth (Bijay-Singh et al., 2008). In addition, a previous study conducted at our site shows that liming increased nutrient availability (Liao et al., 2018), which may have contributed to increased plant N uptake and rice growth.

Liming reduced CH_4 emissions in the second year of our study to a smaller extent than in the first year. One possible explanation for this result would be that liming-induced increases in aboveground biomass in the first late rice season provided additional substrate to methanogens in the next year; this response would partly negate the negative liming effects on CH_4 emissions discussed above. Indeed, numerous studies (e.g. Das and Adhya, 2014; Liu et al., 2014; Sander et al., 2014) show that rice straw incorporation stimulates CH_4 emissions. On the other hand, our findings indicate that increased biomass with liming increased soil C contents; taking this effect into account would actually increase our estimates of GHG mitigation with liming (e.g. Fornara et al., 2011). Clearly, liming affects the GHG budget of rice systems through several processes that operate on different time scales. Thus, long-term experiments are needed to confirm whether the GHG mitigation effects of liming found in our study persist over time.

In conclusion, we found that liming reduces CH_4 emissions from an acid rice paddy soil. Although the exact mechanisms underlying this result are not yet fully clear, there are strong indications that liming reduces methanogenic activity during the rice growing season by increasing litter decomposition during the fallow period, thereby reducing the amount of substrate that is available to methanogens. Based on China's second state soil survey completed in the early 1980s, approximately 32% of Chinese rice paddies have a pH below 5.5 (State Soil Survey Service of China (SSSC), 1993, 1994a,b, 1995a,b, 1996). Moreover, pH of China's paddy soils has reduced by about 8.9% from 1988 to 2013 (Zhou et al., 2015). Thus, our results suggest that liming could play an important role in mitigating greenhouse gas emissions from rice agriculture.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.geoderma.2018.03.034.

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